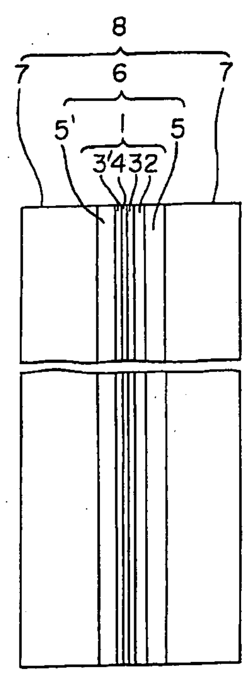
(12) UK Patent Application (19) GB (11) 2 057 355 A

(21) Application No 8027837
(22) Date of filing 28 Aug 1980
(30) Priority data
(31) 54/108620
(32) 28 Aug 1979
(33) Japan (JP)
(43) Application published
1 Apr 1981
(51) INT CL^3
B32B 17/10 // 27/06 27/08
27/30 27/36
(52) Domestic classification
B5N 1710 2706 2708 2730
2736
(56) Documents cited
None
(58) Field of search
B5N
(71) Applicants
Asahi Glass Company
Limited,
No. 1-2 Marunouchi
2-chome,
Chiyoda-ku,
Tokyo,
Japan.
Honda Giken Kogyo
Kabushiki Kaisha,
27-8, 6-chome,
Jingumae,
Shibuya-ku,
Tokyo,
Japan.
(72) Inventors
Yoshitugu Fujimori,
Masataka Kumata,
Mamoru Mizuhashi.
(74) Agents
R.G.C. Jenkins & Co.,
Chancery House,
53/64 Chancery Lane,
London,
WC2A 1QU.

(54) Infra-red reflecting glass laminate, particularly for automobile windscreens

(57) An infrared reflecting glass laminate having at least 70% visible ray transmission is formed by a pair of glass sheets (7) with a flexible laminate (6) of interlayer films between them. This comprises a pair of polyvinyl butyral interlayer films (5,5') at least one of which includes an ultraviolet ray absorbent preferably in an amount of 0.5 to 3 wt.% based on the film. Between the polyvinyl butyral films is a plastics film (2) coated sequentially with a tungsten oxide layer (3), a silver layer (4) and a tungsten oxide layer (3') to form an infrared reflecting film (1).

FIG. 1



GB 2 057 355 A

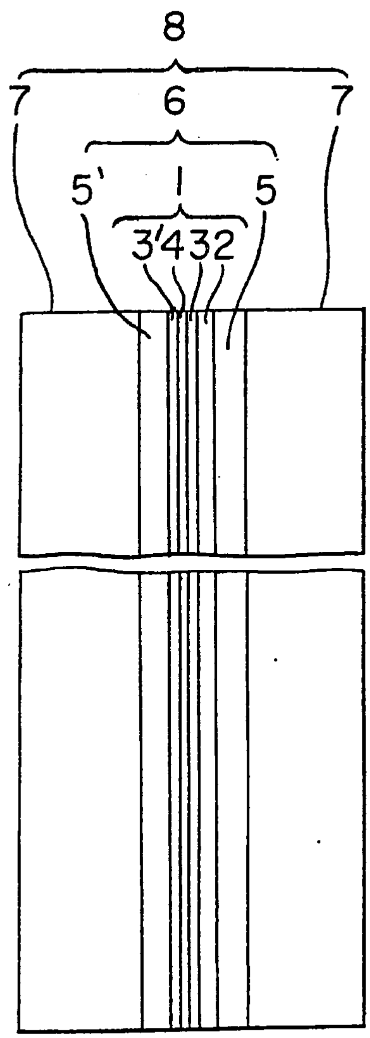
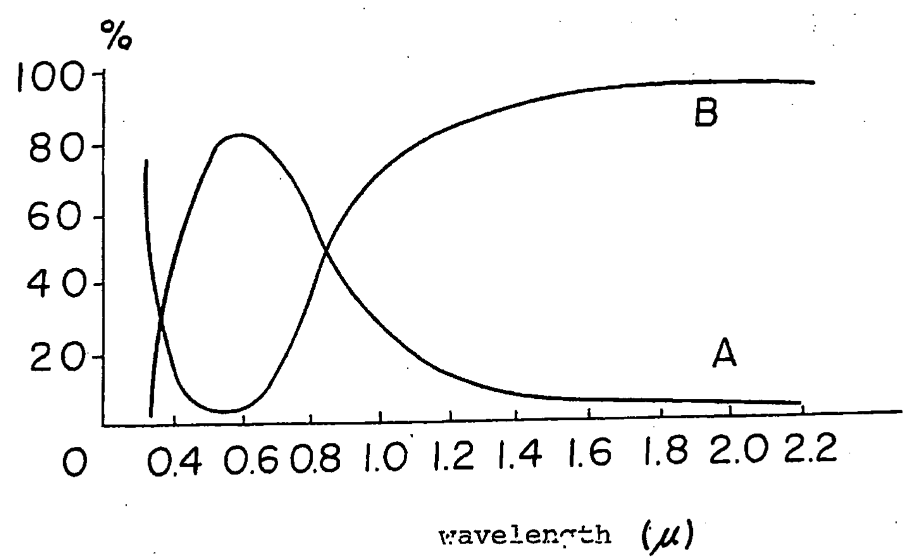


FIG. 1

FIG. 2

A) percentage transmission
B) percentage reflectivity



SPECIFICATION

Infra-red reflecting glass laminate, particularly for automobile windscreens

The present invention relates to an infrared reflecting laminated glass for automobiles.

It is known to use an infrared reflecting glass sheet for a window of a vehicle or a building. This reflects the infrared rays in sunlight on its surface to intercept the transmission of the infrared rays, thereby preventing a temperature rise in the room or decreasing the energy load required for cooling the room. It is known that such infrared reflecting glass can be formed by coating a metal layer made of gold, silver, copper or aluminum on a surface of a glass sheet or coating the surface with a metal oxide layer such as cobalt oxide, iron oxide, chromium oxide, titanium oxide or tin oxide. Such sheets have hitherto been thought unsuitable for an automobile because of its low visible ray transmission or high visible ray reflectivity. It is possible to provide a thin metal layer or metal oxide layer for increasing the visible ray percent transmission, but this has the disadvantage that satisfactory infrared reflecting characteristics are not achieved. For an automobile windscreen, at least 70% visible ray transmission is required by law. It is desirable for the glass to have less than 12% visible ray reflectivity and less than 65% total transmission of solar rays and to intercept ultraviolet rays of wavelength less than 3900Å. It has proved difficult to obtain an infrared reflecting laminated glass which has all these characteristics. There is no regulation for visible ray reflectivity or total solar radiation transmission. According to various experiments conducted by the inventors, when the visible ray reflectivity is more than 12%, the visible ray transmission is too low for an automobile windscreen and moreover, reflected images adversely affect persons around the automobile. When the total solar radiation transmission is more than 65%, overheating inside the automobile cannot be adequately controlled. Deterioration of the interior fittings in the automobile due to ultraviolet rays can be prevented by intercepting ultraviolet rays having a wavelength less than 3900Å.

The inventors have proposed an infrared reflecting glass laminate which is prepared by producing an infrared reflecting plastic film formed by sequentially coating on a plastics film a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å, a silver layer having a thickness of 80 to 150Å and a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å and holding said infrared reflecting plastics film between a pair of polyvinyl butyral interlayer films and laminating a pair of glass sheets with the interlayer films between them. According to various tests of the durabilities of the infrared reflecting laminated glass, the capacity of the laminate to intercept ultraviolet rays is insufficient and the infrared reflecting plastics film is disadvantageously coloured.

The present invention provides an infrared reflecting glass laminate having visible ray transmission of at least 70% which comprises a pair of glass sheets having between them a flexible laminate of interlayer films comprising a pair of polyvinyl butyral interlayer films which hold between them an infrared reflecting plastics film formed by sequentially coating on a plastics film (1) a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å, (2) a silver layer having a thickness of 80 to 150Å and (3) a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å wherein at least one of said polyvinyl butyral interlayer films includes an ultraviolet absorbent.

The laminate of the present invention overcomes the above-mentioned disadvantages and has satisfactorily high infrared ray reflectivity, high visible ray transmission and low visible ray reflectivity and relatively low total solar radiation transmission it is disadvantageously not colored. It is therefore quite suitable for use as an automobile windscreen.

Preferred embodiments of the invention will now be described with reference to the accompanying drawings wherein:

Figure 1 is a schematic sectional view of one embodiment of an infrared reflecting glass laminate in accordance with the present invention; and

Figure 2 show the optical characteristic of the infrared reflecting glass laminate of the invention.

The plastics film on which are coated the infrared reflecting layers used in the laminate of the present invention is preferably a polyester film such as a polyethyleneterephthalate film which has high transparency and flexibility, is not easily broken, is easily processed, can be easily and continuously coated with infrared reflecting layers at high speed and has high weathering resistance and chemical stability. The most preferred film is a plastics film having more than 80% visible ray transmission and a thickness of 25 to 125μ.

The infrared reflecting layers formed on the plastics film are selected so as to meet the regulation for an automobile windscreen which require more than 70% visible ray transmission. The infrared reflecting film has a three layer structure formed by sequentially coating a tungsten oxide layer, a silver layer and a tungsten oxide layer which have respectively desired thicknesses. Among the infrared reflecting layers, the silver middle layer is selected because it has high infrared reflecting characteristics and optimum reflection hue and transmission hue for a windscreen for an automobile. The silver layer comprises silver as a major component but can be modified by incorporating another metal such as Cr, Ni, Al and Ti in an amount of less than 10% or another additive to improve its optical, chemical or mechanical characteristics.

A thickness of the silver layer is preferably in a range of 80 to 150Å so as to provide at least 70% visible ray transmission, less than 12% visible ray reflectivity and a satisfactory infrared ray reflectivity.

In the structure of the infrared reflecting layers, reflection reducing layers sandwich the silver layer so as to

prevent reduction of the percentage visible ray transmission of the silver layer. The reflection reducing layers can be various non-absorbing dielectrics having a reflective index of at least 2. The tungsten oxide layer is selected for the reflection reducing layers in the laminate of the present invention because of its high visible ray transmission and high reflection reducing effect. excellent hue, excellent adhesiveness to the silver layer and the surface of the glass sheet and desirable chemical, mechanical and optical characteristics. The tungsten oxide layer comprises tungsten oxide as its major component but can incorporate up to 10% of another additive.

The thickness of each tungsten oxide layer is in a range of 180 to 500Å or 1400 to 1600Å to give the desired low reflection, high visible ray transmission and hue.

An infrared ray reflecting laminated glass of 5 mm thickness comprising a pair of polyvinyl butyral films holding an infrared reflecting plastics film coated with only a silver layer having a thickness of 180 to 500Å, has a visible ray percent transmission of about 20 to 40%. This is not suitable for the windscreen of an automobile which requires a visible ray transmission of at least 70%.

In the infrared ray reflecting layers having the three layer structure including reflection reducing layers, the percentage visible ray transmission is remarkably increased by photointerference. For example, a laminate made of two glass sheets each having a thickness of 2.5 mm usually has a visible ray transmission of about 70 to 75%. The percentage visible ray reflectivity can be reduced to less than 10% and reflectivities in the infrared region can be maintained at high levels for example, 65% at 0.9μ wavelength; 70% at 1.0μ wavelength and 90% at 1.5μ-wavelength. A vacuum evaporation coating method is usually employed in the formation of the infrared reflecting layers having the three layer structure on the surface of the film of the present invention.

One example of the vacuum evaporation coating method will be illustrated.

In a vacuum tank in 1 to 5 × 10^{-5} torr, each of the evaporation coating materials is heated to deposit the tungsten oxide layer, the silver layer and the tungsten oxide layer in each desired thickness, on a polyethyleneterephthalate film heated at about 20 to 80°C. The silver and tungsten oxide layers can be also coated by a spattering method or an ion plating method instead of the vacuum evaporation coating method. It is also possible to form the silver layer by a chemical plating method and to form the tungsten oxide layer by C.V.D. method or C.L.D. method.

When the coated layers of the infrared reflecting plastic film are exposed to the atmosphere, a chemical durability and a mechanical durability are inferior. In order to overcome the disadvantage and to make easy the lamination of a pair of glass sheets, the infrared reflecting plastic film is sandwiched between a pair of polyvinyl butyral interlayer films. The flexible laminated films are hold between a pair of glass sheets to press-bond them.

When two glass sheets having each thickness of about 2 to 5 mm are laminated, a visible ray percent transmission of the laminated glass is decreased for about 5 to 15%. In order to provide more than 70% of a visible ray percent transmission, it is preferable to provide about 80 to 85% of a visible ray percent transmission of the infrared reflecting plastic film having the infrared reflecting layers in three layer structure.

When two glass sheets having each thickness of about 2 to 5 mm are laminated, a visible ray percent transmission of the laminated glass is further decreased for about 5%. In order to provide more than 70% of a visible ray percent transmission, it is preferable to use each glass sheet having a visible ray percent transmission ranging from 85 to 90%. It is preferable to use each glass sheet having a thickness ranging from about 1.5 to 3 mm for the laminated glass so as to be a small loss of a visible ray percent transmission.

In the present invention, the polyvinyl butyral interlayer films 0.3 - 0.7 mm thickness sandwiching the infrared reflecting plastic film are prepared by incorporating an ultraviolet absorbent so as to intercept ultraviolet rays especially ultraviolet rays having wavelengths of less than 3900Å. The ultraviolet absorbents for plastics are compounds having large ultraviolet absorption: benzophenones such as 2,4 dihydroxybenzophenone; 2-hydroxy-4-methoxybenzophenone; 2-hydroxy-4-n-octoxybenzophenone; 4-dodecyloxy-2-hydroxybenzophenone; 2-hydroxy-4-octadecyloxybenzophenone; 2,2′ dihydroxy-4-methoxybenzophenone; 2,2′ dihydroxy-4,4′ dimethoxybenzophenone; 2,2′ dihydroxy-4-methoxybenzophenone; 2,2′, 4,4′ tetrahydroxybenzophenone; 2-hydroxy-4-methoxy-5-sulfobenzophenone; 2-hydroxy-4-methoxy-2′-carboxybenzophenone; 2,2′ dihydroxy-4, 4′ dimethoxy-5-sulfobenzophenone; 2-hydroxy-4-(2-hydroxy-3-methyl aryloxy) propoxybenzophenone; and 2-hydroxy-4-chlorobenzopheone;

benzotriazoles such as 2(2′ hydroxy-5-methylphenyl) benzotriazole; 2(2′-hydroxy-3′,5′-ditert-butyl phenyl) benzotriazole; and 2(2′ hydroxy-3′-tert, butyl-5′-methyl-phenyl) benzotriazole;

salicylates such as phenyl salicylate; carboxyphenyl salicylate; p-octylphenyl salicylate; strontium salicylate; p-tert, butylphenyl salicilate; methyl salicylate; and dodecyl salicylate;

and also other ultraviolet absorbents such as

resorcinol monobenzoate; 2′ ethyl hexyl-2-cyano; 3-phenylcinnamate; 2-ethyl-hexyl-2-cyano-3,3-diphenyl acrylate;

ethyl-2-cyano-3,3-diphenyl acrylate;

[2-2′-thiobis(4-t-octyl phenolate)]-n-butylamine nickel;

asymmetric oxalic acid diaryl amido (A-NH-Co-Co-NH-B);

aromatic dihydric compound such as the ester derivatives of 4,4-bis(4′-hydroxy phenyl) pentanoic acid;

oxyganosilicon compounds, ex.

$$R^4-R^3-\overset{\overset{\displaystyle O}{\|}}{C}-O-R^2-\underset{\underset{\displaystyle R^1n}{|}}{Si}X_{3-n}$$

$$R^4-R^3-\overset{\overset{\displaystyle O}{\|}}{C}-O-R^2-\underset{\underset{\displaystyle R^1n}{|}}{Si}O_{3-n}$$

(R^1: monovalent hydrocarbon radical,
R^2: divalent hydrocarbon radical,
R^3: divalent hydrocarbon radical).

The ultraviolet absorbent is preferably incorporated at a ratio of 0.5 to 5 wt. % based on the polyvinyl butyral interlayer film, for example, the ultraviolet absorbent is incorporated at a ratio of 1 to 5 wt. % for a polyvinyl butyral interlayer film having a thickness of 0.3 mm and at a ratio of 0.5 to 3 wt. % for a polyvinyl butyral interlayer film having a thickness of 0.7 mm.

The ultraviolet absorbing polyvinyl butyral interlayer film is preferably used for each of the polyvinyl butyral films used in both sides of the infrared reflecting plastic film. In some case, it is possible to use the ultraviolet absorbing polyvinyl butyral interlayer film only for the outer or inner polyvinyl butyral interlayer film for an automobile. When the ultraviolet absorbent is incorporated in the polyvinyl butyral interlayer film, the molecule of the polyvinyl butyral can be stabilized by the oxidation inhibiting effect and ultraviolet ray absorbing effect of the ultraviolet absorbent, whereby a humidity resistance can be improved to increase the durability of the infrared reflecting plastic film.

The ultraviolet absorbent in the polyvinyl butyral affect to the tungsten oxide layer so as to prevent the deterioration of the tungsten oxide layer. This is the significant effect of the combination of the ultraviolet absorbent, polyvinyl butyral and tungsten oxide layer. It is preferable to use the ultraviolet absorbing polyvinyl butyral interlayer film so as to contact with the tungsten oxide layer.

Incidentally, it is also possible to incorporate a ultraviolet absorbent in the polyethyleneterephthalate film on which tungsten oxide layer is formed. The kind and quantity of the ultraviolet absorbent are the same.

Figure 1 shows schematic sectional view of the infrared reflecting laminated glass for an automobile as one embodiment of the present invention. The infrared reflecting plastic film (1) comprises a polyethyleneterephthalate film (2), coated with a tungsten oxide layer (3), a silver layer (4) and a tungsten oxide layer (3'). The infrared reflecting plastic film (1) is sandwiched between a pair of polyvinyl butyral interlayer films (5), (5'). The flexible laminated films (6) are sandwiched between a pair of glass sheets (7), (7') to form the infrared reflecting laminated glass (8).

In the embodiment shown in Figure 1 the infrared reflecting plastic film (1) is sandwiched between pair of polyvinyl butyral films. It is also possible to superpose one or more polyvinyl butyral film to the flexible laminated films.

The present invention will be further illustrated by certain examples which are provided for purposes of illustration only.

Example 1

A polyethyleneterephthalate film (a thickness of 50μ; a length of 50 cm and a width of 100 cm) was washed and dried and placed in a vacuum tank of a vacuum evaporation coating apparatus. The vacuum tank was evacuated to a vacuum degree of 10^{-5} torr and the film was kept at 50°C. Two boats for evaporating sources were placed in the vacuum tank and a silver rod and tungsten oxide powder were respectively charged in each boat. The tungsten oxide was heated at 1200°C to coat a tungsten oxide layer at a deposition speed of 10 Å/sec. whereby a tungsten oxide layer having 350 Å of a thickness was formed on the polyethyleneterephthalate film.

The silver was heated at 1400°C to coat a silver layer at a deposition speed of 20 Å/sec. whereby a silver layer having 100 Å is formed on the tungsten oxide layer having 350 Å of a thickness.

A tungsten oxide layer having 350 Å of a thickness is further coated on the silver layer by the same method. The resulting infrared reflecting plastic film was sandwiched between a pair of ultraviolet absorbing polyvinyl butyral films having each thickness of 30 mil. (Therflex XA manufactured by Mitsubishi Monsanto Co.) The laminated films were further held between a pair of glass sheets (a thickness of 2.5 mm; a length of 50 cm; a width of 100 cm) and laminated by the conventional heat-press bonding method. The polyvinyl butyral film comprised benztriazole at a ratio of 3 wt.%.

Figure 2 shows a spectral transmittance curve and a spectral reflectance curve of the resulting infrared reflecting laminated glass.

According to a boiling test, a natural exposing tests, a weather meter test, it was confirmed that the

infrared reflecting laminated glass had enough durability in a practical use.

CLAIMS

1) An infrared reflecting glass laminate having visible ray transmission of at least 70% which comprises a pair of glass sheets having between them a flexible laminate of interlayer films comprising a pair of polyvinyl butyral interlayer films which hold between them an infrared reflecting plastics film formed by sequentially coating on a plastics film (i) a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å, (2) a silver layer having a thickness of 80 to 150Å and (3) a tungsten oxide layer having a thickness of 180 to 500Å or 1400 to 1600Å, wherein at least one of said polyvinyl butyral interlayer films includes an ultraviolet absorbent.

2) An infrared reflecting glass laminate according to Claim 1 which has a visible ray reflectivity of less than 12%.

3) An infrared reflecting glass laminate according to Claim 1 or Claim 2 which has a total solar radiation transmission of less than 65%.

4) An infrared reflecting glass laminate according to any preceding claim wherein said polyvinyl butyral interlayer film having ultraviolet ray absorbancy intercepts more than 90% of ultraviolet rays having wavelengths of less than 3900Å.

5) An infrared reflecting glass laminate according to any preceding claim wherein said polyvinyl butyral interlayer film comprises said ultraviolet ray absorbent in an amount of 0.5 to 3 wt. % based on said film.

7) A infrared reflecting glass laminate substantially as herein described with reference to the accompanying drawings.

Printed for Her Majesty's Stationery Office, by Croydon Printing Company Limited, Croydon, Surrey, 1981.
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.